

## Dynamics of Axial Separation in Long Rotating Drums

I. S. Aranson

*Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439*

L. S. Tsimring

*Institute for Nonlinear Science, University of California, San Diego, La Jolla, California 92093-0402*

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We propose a continuum description for the axial separation of granular materials in a long rotating drum. The model, operating with two local variables, concentration difference and the dynamic angle of repose, describes both initial transient traveling wave dynamics and long-term segregation of the binary mixture. Segregation proceeds through ultraslow logarithmic coarsening. [S0031-9007(99)09348-5]

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The collective dynamics of granular materials recently have been attracting much interest [1–7]. The dissipative nature of interaction among macroscopic particles sets granular matter apart from familiar gaseous, liquid, or solid states. One of the most fascinating features of heterogeneous granular materials is their ability to segregate under external agitation instead of mixing as would be expected from thermodynamics. Essentially any variation in mechanical properties of particles (size, shape, density, surface roughness) may lead to their segregation.

Granular flow in slowly rotating drums is different from conventional fluid motion. In the bulk, particles perform rigid body rotation around the drum axis until they reach the free surface. Then they slide down within a thin near-surface layer [4]. For intermediate rotation speed, the surface has a rather flat S-curved shape, and arctangent of its average slope defines the *dynamic angle of repose*. Since there is almost no interparticle motion in the bulk, segregation of binary granular mixtures predominantly occurs within the fluidized near-surface layer [3,5,7]. In *radial* segregation grains of one type (for grains of different sizes, the smaller ones) build up a core near the axis of rotation. It occurs during the first few revolutions of the drum. For long drums, along with radial segregation, *axial* segregation occurs at much later stages (after hundreds of revolutions). Recent experiments [3,6,7] revealed interesting features of axial segregation. For rapid rotation speed and large composition fraction of salt grains in a salt-sand mixture  $\phi > 0.55$ , small-scale perturbations travel across the drum at early stages of segregation. At later times longer-scale perturbations take over and lead to quasistationary bands of segregated materials which exhibit slow coarsening [5] leading sometimes to complete segregation [8].

Most of the theoretical models of segregation agree in that the reason for segregation is the sensitive dependence of the surface slope or shape on the relative concentration of different particles in the mixture. In Ref. [6] a simple theory of segregation in thin surface flow driven by the local slope was proposed. For a monodisperse material, the model recovers the S shape of the free surface. For

a binary mixture the model yields a nonlinear diffusion equation for the relative concentration of the ingredients along the horizontal axis. Axial segregation occurs when the diffusion coefficient turns negative. This model yields a significant insight into the nature of the instability leading to the segregation; however, it fails to describe the transient waves at the early stages [7].

In this Letter we propose a continuum model which describes consistently the early phase of segregation with traveling waves as well as the later stage of segregation characterized by slow merging of bands of different particles. Our model predicts slow (logarithmic) coarsening of the segregated state. The dynamics of segregation shows striking similarity with the experiments of Ref. [7].

Let us consider a mixture of two sorts of particles, *A* and *B*, of which *A* (*B*) corresponds to particles with larger (smaller) static repose angle. Our model operates with two variables: the relative concentration of particles  $c = (c_A - c_B)/\langle c \rangle$ , and local dynamic repose angle  $\theta$ . Here  $c_{A,B}$  are local partial concentrations of particles integrated over drum cross section, and  $\langle c \rangle = \langle c_A + c_B \rangle$  is an average concentration over the whole system. We assume that  $c$  and  $\theta$  are functions of longitudinal coordinate  $x$  and time  $t$ . We assume that all parameters and variables are normalized by the radius of the drum and gravity acceleration.

The first equation represents conservation of the relative concentration  $c$  in the binary mixture:

$$\partial_t c = -\partial_x[-D(c)\partial_x c + g(c)\partial_x \theta]. \quad (1)$$

The first term describes diffusion flux, and the second term describes differential flux of grains due to the gradient of dynamic repose angle. The transport coefficients  $D, g$  in general depend on the relative concentration  $c$ . The specific form of these coefficients can be derived from the equation for granular transport in the bulk and surface layer. Flux balance calculation [11] yields the following expressions  $D(c) = D_0(1 - \eta c)$  and  $g(c) = G_0(1 - c^2)$  (the latter was first derived in Ref. [6]). Constants  $\eta$  and  $G_0$  depend on the physical properties of the grains. The constant  $G_0$  can be eliminated by rescaling of

distance  $x \rightarrow x/\sqrt{G_0}$ . The plus sign before the last term means that the particles with the larger static repose angle are driven towards the greater dynamic repose angle. As will be shown below, this differential flux gives rise to the segregation instability. Since this segregation flux vanishes with  $g(c)$  for  $|c| \rightarrow 1$  (which correspond to pure  $A$  or  $B$  states), it provides a natural saturation mechanism for the segregation instability.

The second equation describes the dynamics of  $\theta$ ,

$$\partial_t \theta = \alpha[\Omega - \theta + f(c)] + D_\theta \partial_{xx} \theta + \gamma \partial_{xx} c. \quad (2)$$

Here  $\Omega$  is the angular velocity of the drum rotation, and  $f(c)$  is the static angle of repose which depends on the relative concentration [9]. The constant  $\alpha$  establishes the time scale for the axial segregation. It characterizes the ratio of the number of particles flowing within the thin surface layer to the number of particles advected within the bulk of the drum. In slowly rotating drums most of the particles are brought to the surface via rigid body rotation in the bulk, resulting in  $\alpha \sim 1$ . In contrast, for rapidly rotating drums most of the particles are advected by closed trajectories by the bulk flow, greatly reducing the value of  $\alpha$ .

According to our definition of  $c$ ,  $f(c)$  is an increasing function of relative concentration. Since the angle of repose as a function of the concentration  $c$  changes typically in a small range [10], we can approximate the function  $f(c)$  by linear dependence  $f(c) = F + f_0 c$ . The constants  $\Omega, F$  can be eliminated by the substitution  $\theta \rightarrow \theta - \theta_0$ , where  $\theta_0 = (\Omega + F)$  has the meaning of the stationary dynamic repose angle, which in the limit  $\Omega \rightarrow 0$  approaches the static repose angle. The first term in the right-hand side of Eq. (2) describes the local dynamics of the repose angle ( $\Omega$  increases the angle, and  $-\theta + f(c)$  describes the equilibrating effect of the surface flow), and the term  $D_\theta \partial_{xx} \theta$  describes axial diffusive relaxation. The last term,  $\gamma \partial_{xx} c$ , represents the lowest-order nonlocal contribution from the inhomogeneous distribution of  $c$ . It appears as a result of an interplay between the bulk flow and Fick diffusion [11]. As we will see later, this term gives rise to the transient oscillatory dynamics of the binary mixture.

Let us consider the stability of the uniform state,  $c = c_0$ ;  $\theta_0 = f_0 c_0$ , where  $c_0$  is determined by initial conditions:

$$\begin{aligned} c &= c_0 + C e^{\lambda t + i k x}, \\ \theta &= \theta_0 + \Phi e^{\lambda t + i k x}. \end{aligned} \quad (3)$$

To linearize the system, we need to expand the functions  $D(c), g(c)$  near  $c_0$ . The stability properties depend on the values of  $f_0$  and  $g_0 \equiv g(c_0)$ ,  $d_0 = D_0(1 - \eta c_0)$ . The eigenvalues  $\lambda_{1,2}$  are found from the following equation:

$$(\lambda + d_0 k^2)(\lambda + D_\theta k^2 + \alpha) - g_0(\alpha f_0 k^2 - \gamma k^4) = 0. \quad (4)$$

At  $k \rightarrow 0$  one finds  $\lambda_1 = -\alpha - (D_\theta + g_0 f_0)k^2$ ;  $\lambda_2 = (g_0 f_0 - d_0)k^2$ . Asymptotic expansion at  $k \rightarrow \infty$  yields

$$\lambda_{1,2} = \frac{1}{2} [-D_\theta - d_0 \pm \sqrt{(D_\theta - d_0)^2 - 4g_0 \gamma}] k^2. \quad (5)$$

It is easy to see that if  $g_0 f_0 > \alpha d_0$ , long-wave perturbations are unstable ( $\lambda_2 > 0$ ), and if  $g_0 \gamma > (D_\theta - d_0)^2/4$ , short-wave perturbations oscillate and decay (eigenvalues  $\lambda_{1,2}$  are complex conjugate with negative real part). The full dispersion curve  $\lambda(k^2)$  for particular values of parameters is plotted in Fig. 1. This curve is consistent with the measurements in Ref. [7]. Indeed, at small wavelengths the perturbations travel, and their phase velocity  $v_{ph} = \text{Im}\lambda/k$  asymptotically grows linearly with  $k$ . At  $k = k_*$ , frequency and phase velocity turn into zero and remain zero for all long-wave perturbations  $k < k_*$ . Perturbations in the range  $0 < k < k_c$ , where  $k_c \equiv [\alpha(g_0 f_0 - d_0)/(g_0 \gamma + d_0 D_\theta)]^{1/2} < k_*$ , grow exponentially and there is an optimal wave number of the fastest growing perturbations  $k_0$ . Oscillating perturbations with  $k > k_*$  always decay, which seems to be in agreement with experimental data, although direct experimental measurement of  $\text{Re}\lambda(k)$  is lacking. The explicit dependence of the coefficients  $d_0, g_0$  on  $c_0$  yields the threshold  $c_{min}$  for the transient oscillatory behavior. This threshold was indeed observed in the experiment [7]. Since  $g_0 \rightarrow 0$  as  $c_0 \rightarrow 1$ , there also is an upper limit  $c_{max}$  for the transient oscillations according to our theory. This upper threshold has not yet been observed.

We performed numerical simulations of Eqs. (1) and (2) using pseudospectral split-step method with periodic boundary conditions. We used up to 512 mesh points in our numerical procedure. The following form of nonlinear functions in Eqs. (1) and (2) was implemented:  $g(c) = 1 - c^2$  and  $f(c) = f_0 c$  [12]. For simplicity we set  $\eta = 0$ , so  $d_0 = D_0$ . We also verified that  $\eta \neq 0$  affects only quantitative features (i.e.,  $k^*$ ) without

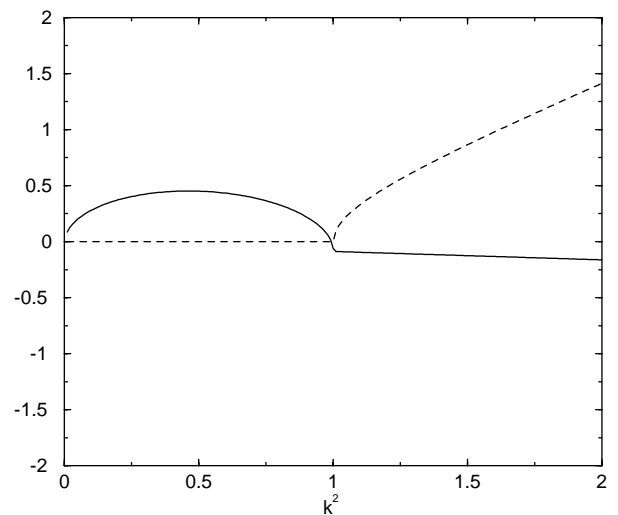


FIG. 1. Dispersion relation  $\lambda(k)$  for the small perturbation of the uniform state  $c_0 = 0$  at  $f_0 = 40$ ,  $D = 0.05$ ,  $D_\theta = 0.1$ ,  $\alpha = 0.025$ ,  $\gamma = 1$ . Perturbations are unstable at  $k < k_c = 0.994$  and oscillate (and decay) at  $k > k_* = 1$ .

changing the qualitative behavior. Comparing the results of our linear stability analysis with the experimental data of Ref. [7] we obtained the representative values of the model parameters:  $\alpha \sim 10^{-3}-10^{-2}$ ,  $f_0 \sim 10-40$ ,  $\eta \sim 1$ ,  $D_\theta/D_0 \sim 1$ ,  $\gamma/D_\theta \sim 10$  [11].

The dynamics of the initially pre-separated state with wave number  $k > k_*$  in a system with size  $L = 60$  is shown in Fig. 2. Short-wave initial perturbations produce decaying standing waves, which later are replaced by quasi-stationary bands (Fig. 2a). The bands are separated by sharp interfaces which are very weakly attracted to each other. In fact, in simulations with parameters corresponding to Fig. 2a we were not able to detect interface merging at all in a reasonable simulation time. However, at higher rates of diffusion and dissipation, the interaction becomes more significant, and it leads to band merging and overall pattern coarsening (see Fig. 2b). In Fig. 3, we present a number of bands as a function of time for this run.

Fronts separating bands of different grains can be found as stationary solutions of Eqs. (1) and (2). In an infinite system one finds from stationary Eq. (1)  $\theta = \theta_0 + DG(c)$  (we again assume  $\eta = 0$ ), where  $G(c) = \int [g(c)]^{-1} dc = -\frac{1}{2} \ln \frac{1-c}{1+c}$  and  $\theta_0$  is an integration constant. Plugging this expression in Eq. (2), we obtain the second-order differential equation for  $c$  (for a symmetric solution one chooses  $\theta_0 = 0$ ),

$$\frac{d}{dx} \left[ \left( \gamma + \frac{DD_\theta}{1-c^2} \right) \frac{dc}{dx} \right] + \alpha f_0 c + \frac{\alpha D}{2} \ln \frac{1-c}{1+c} = 0. \quad (6)$$

This equation indeed possesses an interface solution. Its asymptotic behavior can be found in the limit  $D \ll f_0$ , when the states on both sides of the interface are well segregated ( $|c(x \rightarrow \infty)| \rightarrow \pm 1$ ). In this limit, far away from the interface  $1 - |c| \propto \exp(-x/l)$ , where

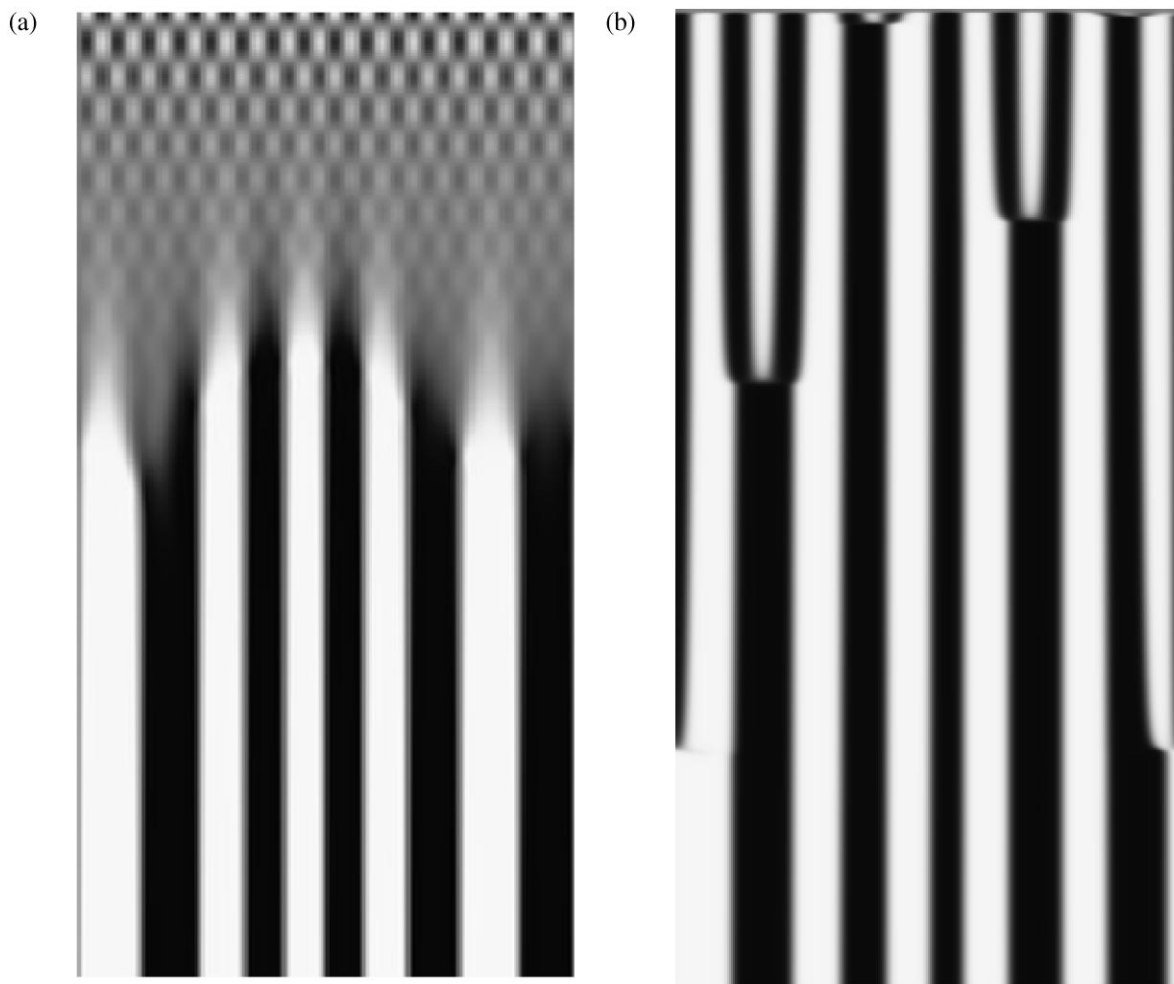


FIG. 2. Space-time diagrams of the evolution of the pre-separated state [ $c(x,0) = c_0 + c_i \cos(k_0 x)$ ] with  $c_0 = 0$ ,  $c_i = 0.95$ , and  $k_0 = 1.79 > k_*$ ; time increases from top to bottom,  $x$  coordinate from left to right. (a) Initial transient. At times  $t < 15$  the initial perturbations excite decaying standing wave (superposition of left- and right-traveling waves), and at larger times  $t > 15$ , aperiodic segregated bands emerge. Parameters of the model are  $D = 0.05$ ,  $\gamma = 1$ ,  $\alpha = 0.025$ ,  $f_0 = 40$ ,  $L = 60$ , and  $D_\theta = 0.1$ . (b) Space-time diagram for long-time evolution, band merging, and coarsening during long-time evolution ( $0 < t < 10000$ ) at higher diffusion constants, parameters  $D = 0.8$ ,  $D_\theta = 0.5$ ,  $\gamma = 1$ ,  $\alpha = 0.5$ ,  $f_0 = 2$ ,  $L = 140$ .

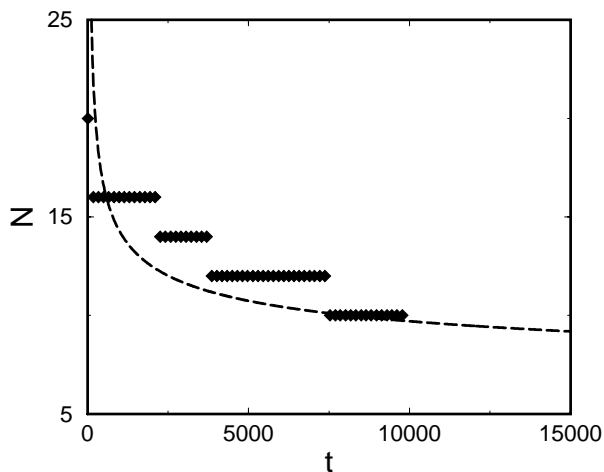


FIG. 3. Number of fronts  $N$  as a function of time (diamonds) and its fit by a function  $N = 70/(\ln t - 2.5)$  (long-dashed line). Parameters correspond to Fig. 2b.

$l = \sqrt{\frac{D_\theta}{\alpha} + \frac{4\gamma}{\alpha D} \exp(-\frac{2f_0}{D})}$ . As seen from this formula, the characteristic front width vanishes as  $D, D_\theta \rightarrow 0$ . This could be anticipated, as in the absence of diffusion nonlinearity  $g(c)$  drives the system towards complete segregation.

Logarithmic coarsening of the segregated state can be understood in terms of the weak interaction of fronts. Since the asymptotic field of the front approaches the equilibrium value of the concentration exponentially fast, we expect exponential times for front interaction,  $T \propto \exp(d/l)$ , where  $d$  is the initial distance between fronts. For multiband structure, the number of fronts  $N$  (proportional to the inverse average distance between fronts) decreases approximately as a logarithmic function of time  $N = 1/d \sim 1/(\text{const} + l \ln T)$ . This dependence indeed agrees with our numerical simulations (see Fig. 3).

In conclusion, we proposed a simple continuum model for axial segregation of binary granular mixtures in long rotating drums. The model operates with two local dynamical variables, relative concentration of two components and dynamic repose angle. The dynamics of our model shows qualitative similarity with the experimental observations of initial transients and long-term segregation dynamics [3,5–7]. It captures both initial transient traveling waves and subsequent onset of the band structure. The dispersion relation for the slightly perturbed uniform state (4) qualitatively agrees well with observations [7] and can serve for fitting the model parameters. More detailed derivation of the model and comparison with experiments will be given elsewhere [11]. The model also describes logarithmic coarsening of the quasistatic band structure. This coarsening is typical for one-dimensional systems with exponentially weak attractive interaction among defects or interfaces, as in the phase ordering kinetics described by the Cahn-Hilliard model [13]. As in the Cahn-Hilliard

model, the order parameter (here concentration  $c$ ) is a conserved quantity; therefore, front interaction must conform to a global constraint. Our simulations also showed that the model qualitatively reproduces more complicated phenomenology of the separation process reported in Ref. [6]. In particular, periodic modulation of the drum radius, modeled in our approach by a periodic variation of  $\Omega$ , leads to band locking. Breaking of the  $x \rightarrow -x$  symmetry results in complete segregation in a much shorter time, similarly to the dynamics of grains in the drum with helicoidal shape. Finally, this model operates with variables averaged over the cross section of the drum and thus cannot describe radial segregation. A more elaborate three-dimensional model is needed to describe both radial and axial segregation within a unified framework.

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- [1] H.M. Jaeger, S.R. Nagel, and R.P. Behringer, *Phys. Today*, **49**, No. 4, 32 (1996); *Rev. Mod. Phys.* **68**, 1259 (1996).
- [2] P. Umbanhowar, F. Melo, and H.L. Swinney, *Nature (London)* **382**, 793 (1996).
- [3] K.M. Hill, A. Caprihan, and J. Kakalios, *Phys. Rev. Lett.* **78**, 50 (1997); K.M. Hill and J. Kakalios, *Phys. Rev. E* **49**, 3610 (1994); **52**, 4393 (1995).
- [4] F. Cantelaube and D. Bideau, *Europhys. Lett.* **30**, 133 (1995); E. Clément, J. Rajchenbach, and J. Duran, *Europhys. Lett.* **30**, 7 (1995).
- [5] V. Frette and J. Stavans, **56**, 6981 (1997).
- [6] O. Zik, D. Levine, S.G. Lipson, S. Shtrikman, and J. Stavans, *Phys. Rev. Lett.* **73**, 644 (1994).
- [7] K. Choo, T.C.A. Molteno, and S.W. Morris, *Phys. Rev. Lett.* **79**, 2975 (1997); K. Choo, M.W. Baker, T.C.A. Molteno, and S.W. Morris, **58**, 6115 (1998).
- [8] R. Chicarro, R. Peralta-Fabi, and R.M. Velasco, in *Powders and Grains '97*, edited by R.P. Behringer and J.T. Jenkins (A.A. Balkema, Rotterdam, 1997), p. 479.
- [9] The repose angle has been treated as a dynamical variable in a description of avalanches by T. Bouteux and P.-G. deGennes, *J. Phys. I (France)* **6**, 1295 (1996).
- [10] K.M. Hill, J. Kakalios, K. Yamane, Y. Tsuji, and A. Caprihan, in *Powders and Grains Conference Proceedings* (Balkema, Rotterdam, Brookfield, 1997).
- [11] I. Aranson, L. Tsimring, and V.M. Vinokur (to be published).
- [12] Nonlinearity in  $f(c)$  can also contribute to the saturation of the segregation instability. However, since measurements [10] show little nonlinearity of  $f(c)$ , the simplest form of  $f(c)$  is sufficient.
- [13] J.W. Cahn and J.E. Hilliard, *J. Chem. Phys.* **28**, 258 (1958).